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ABSTRACT: The present study focuses on short flax fiber, as well as long flax fiber-reinforced polypropylene (flax/PP) composites, manufactured by the injection molding method. Compounding of flax with two different grades of PP (with and without maleic anhydride (MA-PP) grafting) is carried out by four methods: kneading process, Henschel kinetic mixer, extrusion compounding, and production of long fiber thermoplastic (LFT) granules through pultrusion. The effect of the compounding method and injection molding on the fiber length and mechanical properties of the composites is being investigated. Furthermore, the effect of fiber–matrix adhesion on the mechanical response is being discussed. It can be concluded that the reduction in fiber length, associated with injection molding, did not affect the tensile properties significantly for the studied systems due to improvements in fiber orientation along the polymer flow direction and increased fiber efficiency through dimensional changes due to fiber opening. The addition of MA-PP led to improvements in the tensile strength of injection-molded composites. Kneader compounded composites showed maximum tensile strength as well as stiffness when compared with other compounding methods.

KEY WORDS: short fiber composites, flax fibers, PP, LFT, compounding, injection molding, MA-PP, interface.

INTRODUCTION

MOST COMPOSITE PARTS are large parts produced in small quantities; the major exception being injection-molded short-fiber-reinforced composite parts. Injection molding requires high capital investment for equipment and tooling so that only large-scale mass production is economic. Furthermore, it offers the advantage of rapid processing into complex shapes but at the expense of fiber length retention. Compared with compression molding, large injection-molded structures have often lower stiffness and impact strength due to lower fiber loadings and shorter fiber lengths. However, the short fabrication times and reduced finishing operations needed during injection molding lead to

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considerable cost reduction during production. Injection molding accounts for 32% by weight of all polymeric materials processed [1]. Transport, consumer products, electrical and electronic, building and construction applications are the main sectors in which growth is taking place.

Glass fiber (GF) is by far the most widely used reinforcement, with practically all injection molding thermoplastic resins available as glass-reinforced grades and usually supplied as ready-to-mold granular compounds or as bulk material. However, in recent years, natural fibers are aiming at replacing glass in fiber-reinforced composites [2,3]. Depending on the exact quality needed, they are in most cases cheaper than glass fibers [4]. Natural fibers are expected to give less health problems for the people producing the composites, since they do not cause skin irritations and are not suspected of causing lung cancer. They are also expected to cause less wear to screws and tooling since they are not as abrasive as glass fibers. Next to that natural fibers are biodegradable, recyclable, and come from renewable natural resources. Furthermore, composites made out of natural fibers are lightweight and have high specific stiffness compared to those made out of glass fibers [5–9]. They already find use, especially in the automobile industry [10–13].

Since injection molding has been a successful method for mass production of glass-fiber-reinforced plastics (GRP), it is of great interest to optimize this process for the production of geometrically sophisticated structural parts out of natural short-fiber-reinforced polymers. Surprisingly, so far little focus has been given to injection molding with natural fibers. A market study on the use of natural fibers in the automotive industry in Germany [14] stated that most natural fiber composites are being manufactured via compression molding of non-woven fabrics and only 2% of the natural fiber composites are being injection molded. As in the case of short glass fiber thermoplastics, the method of fiber addition to the polymer matrix (compounding) can lead to significant fiber breakage and influence the morphology and final properties of the composite. Next to fiber breakage, compounding may also separate the technical cellulose fibers into elementary fibers [15]. It is well known that in discontinuous fiber-reinforced composites the ineffective use of the intrinsic properties of the reinforcing fiber might be due to: (a) subcritical length of the fibers after processing, (b) poor fiber–matrix adhesion and (c) fiber diameter and fiber properties [16,17]. The higher the tensile strength of the fiber and the better the fiber–matrix adhesion, the shorter the minimum fiber length needed for effective transfer of the stress [17,18]. If the fiber length (L_f) in the composites is shorter than the critical fiber length (L_c), then the fibers will not effectively reinforce the composite. On the other hand, fibers that are much longer than the critical fiber length ($L_f > 5L_c$) will effectively reinforce short-fiber-reinforced composite, with in the case of uniaxial alignment, properties approaching those of continuous-fiber-reinforced composites. This critical fiber length can be predicted based on micromechanical models and is different for stiffness, strength, and impact properties [18–20].

Based on the above, in order to exploit the maximum reinforcing ability of natural fibers, in short-fiber-reinforced composites, it is important to select a strong fiber, as well as optimize the fiber length, and fiber–matrix adhesion. Compared to other natural fibers, flax fibers are relatively strong. For flax, very high tensile strengths have been reported (up to 1500 MPa for elementary fibers and circa 800 MPa for technical fibers, which are basically bundles of elementary fibers) [21,22]. This is thought to be due to the fact that flax has the longest elementary fibers and the lowest microfibril orientation. In addition, average Young's moduli of 60–80 GPa for elementary fibers and 40–60 GPa have been reported [22]. Polypropylene (PP) on the other hand is undoubtedly one of the best

candidates as matrix material for natural fiber-reinforced composites, because of its low price, its low melting temperature below the thermal decomposition temperature of natural fibers [23], the problem-free ecological recycling behavior, and its widespread use in technical applications, e.g. in the automotive industry. However, the key issue for natural fiber-reinforced polymer composites stems out of the fact that there is a phase incompatibility between the hydrophilic natural fibers and the non-polar polyolefin matrix (e.g., PP).

The literature found on flax/PP composites refers mainly to two manufacturing routes, (i) mat technology, where non-woven flax mats are compression molded with PP to produce random natural fiber mat composites (NMTs) [2,5,6,18,22,24–26] and (ii) the granule technology where flax fiber-reinforced PP granules are injection molded or extrusion compression molded (ECM) to produce mainly short-fiber-reinforced composites [17,24,27–33]. Limited work is available on unidirectional and multidirectional flax/PP composites [34], while very recently research is also looking into commingled yarn technology for flax/PP [35]. The effect of fiber length and fiber volume fraction has received substantial interest in NMTs based on flax/PP [2,5,6]. The influence of interface compatibilizer has been also discussed in compression-molded flax/PP, where no significant improvements on the mechanical properties [2,18] were found with the addition of compatibilizers. On the other hand, for injection-molded flax/PP composites, positive effects of coupling agents on final mechanical properties have been reported in numerous studies [18,24,28–33]. These studies conclude that an adhesion promoter based on a MA-PP behaves as a true coupling agent, i.e., improves the mechanical performance of the flax/PP composites [18,28–33]. This behavior is linked with the fiber length in injection-molded composites, where normally shorter fibers are found compared to compression-molded composites. Therefore the fiber–matrix adhesion becomes more critical and has a significant effect when the injection molding technology is followed. Different compounding routes prior to injection molding have been employed, i.e., kinetic mixers, granulators, twin screw extruders, kneaders, etc. However, very little data is available on the effect of compounding and injection molding, on the resulting fiber length and on the performance of the end product for flax/PP composites [31,32]. In the studies of Nystrom et al. [32] and Bos et al. [31], the effect of compounding and injection molding on the fiber length degradation is studied in combination with the mechanical response of the obtained composites and supported by micromechanical modeling. In both cases however, the fiber length distribution is measured after injection molding and it is not clear at which stage, i.e., compounding or injection molding, the most severe degradation happens. Finally, the thermal stability [23,36], environmental durability [25,33,34,37], temperature related properties [24] and effect of transcrystallization on the interfacial and macromechanical properties [38] of flax/PP composites have been of interest in the past.

Based on the above analysis, the first aim of the current study is to evaluate separately the effect of the compounding process and injection molding on the fiber length degradation and fiber length distribution in short and long flax/PP composites. For this, four possible methods for compounding of flax with PP are being employed, i.e., the co-kneader, the Henschel kinetic mixer, the pelletizing press followed by extruder mixing, and a special technique developed for production of long fiber granules (LFT) [39]. The fiber length distribution is measured after compounding and after the injection molding process. Conclusions are drawn on the overall performance of each compounding method on the residual length and mechanical properties of the short and long fiber flax/PP composites. MA-PP is used as the interface compatibilizer and its effect on the mechanical

response of the flax/PP composites is examined. Finally, the mechanical response of injection-molded specimens is being compared with that of NMTs (with and without MA-PP), manufactured by compression molding [2] and data on commercially available glass mat PP (GMT).

EXPERIMENTAL PROCEDURE

Materials

Two different PP grades were used as matrix for manufacturing short and long flax fiber composites, i.e., PP with and without 3 wt.% MA-PP. The basic properties of these materials are presented in Table 1.

Flax fibers were used as reinforcement throughout this study to manufacture the composites. For the short fiber composites, the starting fiber length was 10 mm, while 40 mm was used for the long-fiber-reinforced composites.

Compounding Methods and Injection Molding

For manufacturing short fiber composites, in the case of GF/PP, compounding is accomplished mainly with one of the three different possible types of compounders [40], i.e., (1) single screw extruder, (2) twin screw extruder and (3) co-kneaders. These methods used for the easily free flowing glass fibers, to manufacture short and long fiber composites, do not work for the fluffy flax fibers and new methods have to be developed. In the case of flax fibers, feeding through a hopper leads to fiber agglomeration and entanglements and the feeding is not properly controlled.

Four possible methods for compounding of flax with PP are followed: the kneading process, the Henschel kinetic mixer, the use of a pelletizing press to make fiber pellets followed by extrusion, mixing, and a special technique developed for production of long natural fiber granules [39].

In the kneading process (method A), prior to molding, the polymer is blended with the natural fiber. The mixed compound is then granulated and afterwards injection molded. For the kneading process, a laboratory scale (60 cm^3) Haake Kneader (model-HBI System-90) was used. The compounding was done at 30 rpm for 30 min at 210°C.

When a Henschel kinetic mixer (method B) is used, the fibers and the polymer powder or polymer granules are simply introduced into the mixer together with possible additives, such as coupling agents. The rapid stirring generates heat that causes the polymer to soften and almost melt, at which point the fibers and polymer combine. The hot fiber–polymer mixture is then dumped into a cold mixer in which the material is broken down to small

Table 1. Matrix materials used to compound the flax fiber composites and their basic properties.

Material	Manufacturer	Density	Melt flow index
PP Stamylan 112MN40 (**PP**)	DSM	0.905 g/cm^3	230°C/2.16 kg = 47 g/10 min
Maleic anhydride grafted PP Polybond 3150 (**MA-PP**)	Uniroyal Chemical	0.910 g/cm^3	230°C/2.16 kg = 50 g/10 min

granules. Where there is too much difference in the shape and size of the granules, it is necessary to sieve out the bigger granules. For the present study, a Henschel kinetic mixer FM 200, coupled to a cool mixer KM 350 was used. Flax fibers were compounded with MA-PP blend using the Henschel kinetic mixer. The rotor speed of the mixer was set at 1320 rpm for temperatures up to 166°C and it dropped to 660 rpm for temperatures between 166 and 171°C.

The compressed fiber pellet method, on the other hand, offers the possibility of using an extruder in the compounding step. Granules made in an extruder are more regularly shaped than those made in a Henschel mixer or kneader and therefore cause fewer problems during feeding in a subsequent injection molding process. Not only that, the pelletizing process could be cheaper, more simplified and can be applied for large-scale production. Furthermore, the volume reduction obtained through the fiber pelletizing process fits in with standard compounding operations, which are used to (polymer) pellets. The fiber pellets are afterwards compounded on a twin screw extruder (method C). At the end of the extruder, the melt goes through a die plate, into a waterbath, finally followed by a cutter. The compressed fiber pellets were made in a laboratory pellet press (Amandus Kahl, Hamburg, Ger.). The fiber obtained through this pelletizing press were dried and then compounded on a Werner and Pfleiderer co-rotating twin screw extruder (ZSK 25) with a screw diameter of 25 mm and a length of 1060 mm ($l/d = 42$). The matrix material was fed into the extruder in zone 0 using a K-Tron gravimetric feeder at a rate of 100 g/min. As no suitable gravimetric feeding system was available for the dosing of the flax pills, a volumetric feeding screw was used. This screw fed the pills at a rate of 35–50 g/min to a Werner & Pfleiderer twin screw side feeder (ZSB) connected to the first feeding hole (third element), which transported the pellets into the extruder. No degassing was used and, after cutting, the pellets were dried and sieved to 7 mm. The process parameters used here are illustrated in Table 2.

Finally, the Thüringisches Institut für Textil- und Kunststoff-Forschung (TITK), Rudolstadt-Schwarza, Germany, developed a procedure that produces long fiber thermoplastic (LFT) granules from fiber slivers manufactured through textile preparatory method (method D). The long fiber granules used in this study were kindly provided by TITK and the processing details are described in [39]. The principle of the process involves fiber slivers that consist of a mixture of natural fibers and synthetic thermoplastic fibers. The blended sliver is heated in a preheating zone, pulled through a jet and then twisted. The natural fibers are impregnated with the melting matrix polymer fibers by twisting. A compact material strand is formed that can be hauled off continuously. After twisting, the strand is cooled in the cooling zone. Finally, the strand is cut to make pellets (granules). The manufacturing of a sliver for LFT requires a minimum fiber length of more

Table 2. Overview of the process parameters used for compounding of PP/flax method C.

	Temperature of different zones in the barrel (°C)									Die (°C)	Speed (rpm)	Torque (%)
	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10		
PP	195	200	200	200	200	200	200	200	210	220	100	60

	Screw configuration													
ts	5*t	3*kt1	tneg	3*t	3*kt2	tneg	4*t	kt1	kt2	tneg	3*t	2*tt	2*t

where ts = small transport element 25 mm, t = transport element 36 mm, tt = transport element with teeth 20 mm, tneg = negative transport element, kt1 = kneading element 36 mm, kt2 = kneading element 24 mm.

than 40 mm (or at least twice the cut pellet length). The LFT pellets can be used for injection molding processes. The granule material can be adapted to the specific processing requirements regarding fiber content, fiber length, and material composition.

Injection Molding

For the present study tensile test bars were made in ARBURG 250-75 A 220 D injection molding machine. The test bars were dumbbell shaped with dimensions according to standard ASTM D638M ($150 \times 10 \times 3.5\,mm^3$). Parameters used for the injection molding of flax/PP and flax/MA-PP are given in Table 3.

Testing Methods

FIBER LENGTH DISTRIBUTION

In order to measure the remaining fiber length after compounding and injection molding, fibers were extracted from the compounded material and the length was measured. For this reason, the polymer matrix was dissolved in hot xylene. After dissolution of the matrix, flax fibers were isolated by means of filtration. Pictures of these fibers were taken through a Zeiss M35F light microscope. The fiber length distribution was measured by an image processing system developed at Eindhoven University of Technology, which was used to process the data for 150 fibers.

Mechanical Properties

TENSILE TESTING

Tensile tests were performed on a Frank 81565 tensile testing machine at a cross head speed of 5 mm/min according to ASTM D 638 method. Tensile property evaluations were carried out at temperatures below and above ambient temperature using a temperature cabinet and controller. Samples were conditioned at the test temperature, for 10 min, prior to testing. The dumbbell-shaped samples were stored for at least 24 h before testing.

IMPACT TESTING

Charpy (notched) impact tests were also conducted on injection-molded samples with a width of 12.7 mm and a thickness of 4 mm. The samples were loaded over a span of 44 mm at a speed of 3.5 m/s using a servo-hydraulic testing machine equipped with a Charpy

Table 3. Parameters for injection molding process for flax/PP and flax/MA-PP.

Matrix material	PP/MA-PP	PP
Temp. zone 1 (°C)	200	195
Temp. zone 2 (°C)	210	200
Temp. zone 3 (°C)	215	205
Temp. zone 4 (°C)	220	210
Dosing volume (cc)	20	20
Injection pressure (bar)	750	700
Cooling time (s)	18	25
Temp. of the mold (°C)	40	40

impact test fixture. The impact energy was calculated by dividing the total absorbed energy by the cross-sectional area of the sample behind the notch.

RESULTS AND DISCUSSION

Effect of Compounding and Injection Molding on Fiber Length

As mentioned in the experimental part, four compounding methods were selected to manufacture short and long flax fiber-reinforced composites. Figure 1 shows the granules obtained from the four different compounding methods. From the figure, it is obvious that each compounding method will lead to different degradation of the flax fiber length in the compounds. However, it is not obvious what this distribution of this length is. For this reason, fiber length measurements were performed according to the procedure described in the experimental part.

Figure 2(a) presents the fiber length distribution of the fiber pills obtained through the fiber pelletizing method. Next to that, the fiber length distribution for the compounds obtained through methods B-D is presented (Figure 2(b)–(d)). As seen in this graph, the long fiber granule method (Figure 2(d)) results in maximum fiber length in the fiber granules, since the starting fiber length in this case is also much higher (20 mm) compared

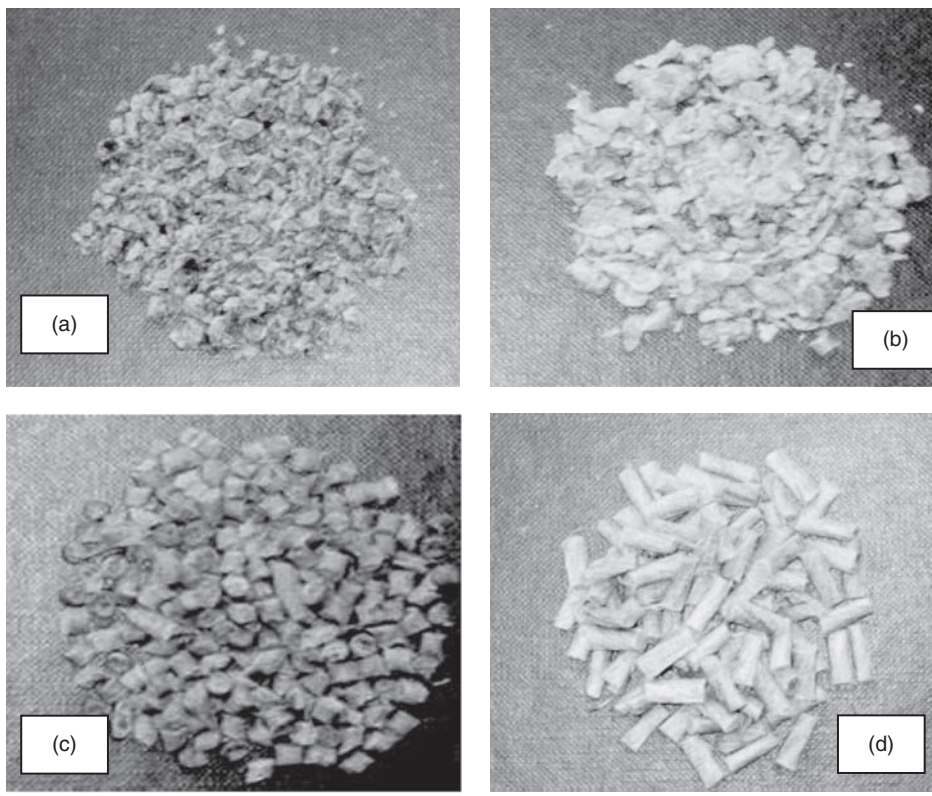

Figure 1. *Picture of granules obtained from various operations: (a) kneader, (b) Henschel mixer, (c) twin screw extruder, and (d) LFT.*

to the other three methods (10 mm). The fiber pelletizing method results in the next highest fiber length (Figure 2(a)). Although longer fibers are obtained after this method, the extrusion compounding process, which follows the fiber pelletizing method leads to more fiber degradation (Figure 2(c)). The resulting fiber length in the twin screw extrusion process is lower than that from the Henschel method (Figure 2(b)). Therefore, overall the Henschel compounding method seems to be better than the pelletizing method followed by twin screw extrusion in retaining the fiber length. On the other hand, the twin screw extrusion method leads to the production of more uniform granules when compared with methods like kneading or mixing, which benefits subsequent feeding processes. Finally, when compared with LFT, the twin screw extrusion method is more simplified, and can be used for large-scale production and probably it is cheaper than the production of LFT, which involves sliver production followed by a (fairly slow) pultrusion process.

In order to find out the fiber length distribution, after compounding and injection molding, fibers were extracted from the molded tensile bars and the fiber length was measured. The obtained results are plotted in Figure 3 for all the compounding methods. On comparing Figure 3 with the fiber length distribution in Figure 2 (before injection molding) it can be stated that injection molding led to additional fiber length reduction in all cases. However, the most significant effect is found in the case of LFT, simply because

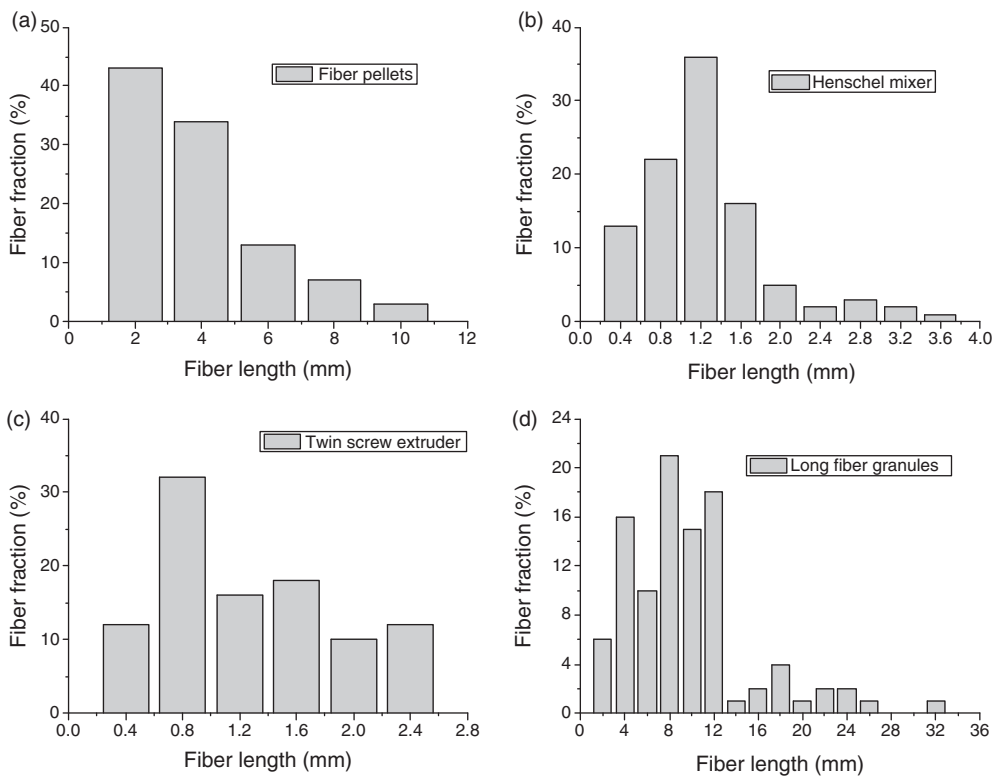

Figure 2. *Fiber length distribution after various compounding methods: (a) fiber pelletizing method, (b) Henschel mixer, (c) twin screw extruder, and (d) LFT.*

the starting length here was highest. The weighted average of the fiber length after compounding and injection molding is 1.2 mm for kneading (range between 0.5 and 4 mm), 1 mm for Henschel compounding (range between 0.4 and 2.4 mm), 0.8 mm for twin screw extrusion (range between 0.4 and 2 mm) and 4.2 mm for LFT (range between 2 and 10 mm). It is obvious that kneading resulted in the highest mean fiber length compared to Henschel mixer and twin screw extruder.

A direct comparison of Figures 2 and 3 is presented in Figure 4. In the case of Henschel compounded granules (Figure 4(a)) the fiber length was already reduced to such an extent that injection molding did not have a significant effect. The maximum fiber length reduced from 3.6 to 2.4 mm and the mean fiber length is slightly shifting to lower values. In the case of the twin screw extrusion method, the effect of the injection molding process on the fiber length is not significant. The maximum fiber length reduced slightly (from 2.4 to 2 mm), since the extrusion process had already introduced the maximum fiber degradation. However, the combination of the injection molding process with twin screw extrusion led to a significant shift of the mean fiber length to lower values. Before injection molding, 88% of the fibers had a length of $\geq$0.8 mm. After injection molding, only 55% of the fibers had a length $\geq$0.8 mm. Finally, in order to compare the effect of kneading and injection molding on the fiber length, a light microscopy image is taken after matrix dissolving. This image is shown in Figure 5 and shows that kneader compounding followed by injection molding reduced the mean fiber length from 10 mm to around 1 mm.

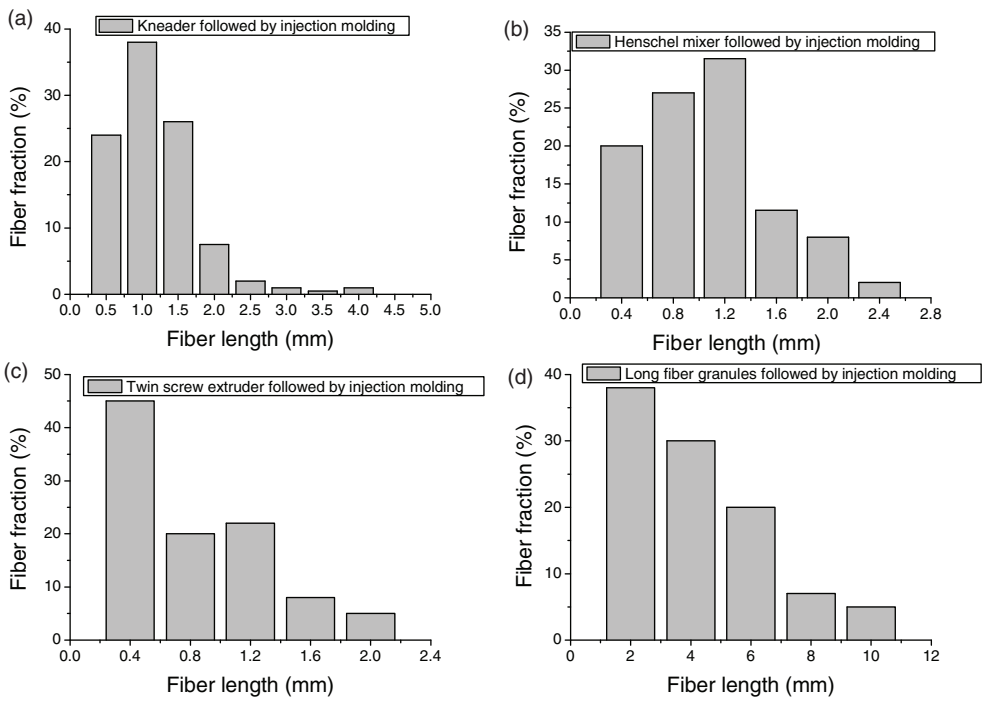

Figure 3. *Fiber length distribution measured after dissolving the matrix in injection-molded composite containing 28 wt.% after various compounding methods: (a) kneader, (b) Henschel mixer, (c) twin screw extruder, and (d) LFT.*

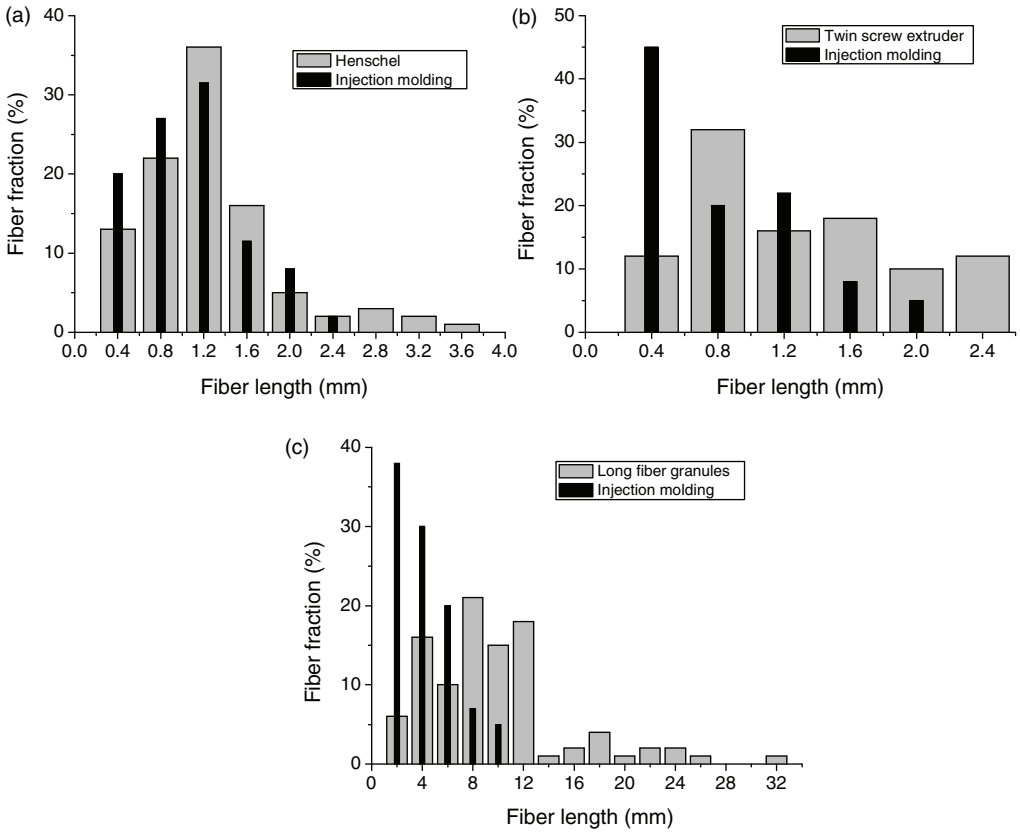

Figure 4. *Direct comparison of the fiber length distribution after compounding and injection molding of the compounded granules for three different compounding methods: (a) Henschel mixer, (b) twin screw extruder, and (c) LFT.*

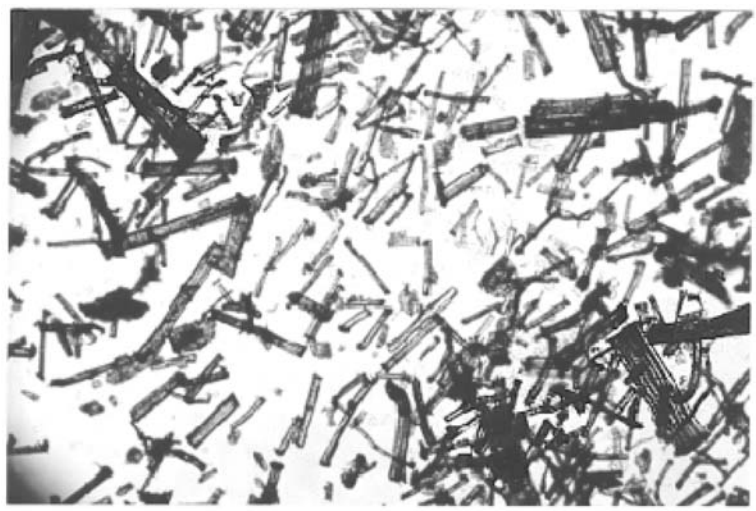

Figure 5. *Picture of fibers after dissolution of matrix polymer in injection molded composite (kneader method).*

Effect of Compounding and Injection Molding on the Mechanical Properties of Flax/PP

The tensile properties of flax/PP composites, compounded through various methods followed by injection molding, are given in Table 4. In this table, the tensile strength, Young's modulus and impact strength is given for the tested composites under different temperatures and flax fiber loadings. From Table 4, it can be concluded that as expected an increase in testing temperature led to a drop in tensile properties. Addition of higher fiber content, in the case of fiber pelletizing followed by twin screw extruder, did not lead to further improvements in the tensile properties. Such behavior could be because of the reduction in fiber length as well as fiber degradation. However, the addition of 3 wt.% MA-PP led to an enhancement in the tensile properties. The composite tensile strength at around 29 MPa was even less than the tensile properties of pure PP (32 MPa), which was compensated for by the addition of coupling agent MA-PP. It can also be concluded that production of composites through LFT led to an increase in impact strength probably due to increased energy absorption through fiber fracture, debonding and pull-out. As such, the impact strength of long fiber composite cannot be compared with other composites, as the composite fracture mode was different. In fact, in the case of long fiber composites, the samples did not break completely after the notched Charpy impact test.

In Figures 6 and 7 a direct comparison of the tensile properties of flax/PP compounds by various techniques followed by injection molding is presented. For reference, the tensile properties of 3 wt.% MA-PP are plotted.

As seen from Figure 6 compounding through kneading led to the highest tensile strength. However, it is worth mentioning here that long fiber composites did not contain any coupling agent and addition of the same is expected to enhance the tensile properties of these composites. The tensile strength of kneader compounded composites was better than that of Henschel compounded as well as twin screw extrusion compounded systems, which was because of longer fiber lengths in the kneader compounded composite as seen from the fiber length distribution (Figure 3). Batches with small quantities were made in a lab-scale kneader and hence this method is not appropriate for large-scale production. Production trials in large scale kneaders have to be done to see the effect of scale on the properties of the composite. Although fibers were longer in the case of LFT, their tensile strength was lower than that of composites made via kneader compounding. This difference could be down to effect of coupling agent, which was not present in the case of the LFT. Finally, it should be noted that compared to unreinforced PP, it seems that the addition of flax fibers in terms of increasing tensile strength was only beneficial when the kneader method was used. However, the other methods did not deteriorate the tensile strength of the flax/PP composites.

Similar to tensile strength the stiffness (Figure 7) of kneader-compounded composites was found to be highest among the composites manufactured. However, it should be noted that here all compounding methods led to stiffness improvements of the composite materials by a factor of 2–3. Such a difference in stiffness cannot be only attributed to longer fiber length retention as well as adhesion enhancement through addition of coupling agent, as in the case of composite strength. The difference could be related to fiber dispersion in the matrix.

In order to investigate the effect of injection rate during injection molding on the tensile strength of flax/PP composite, four injection speeds have been used for a flax/PP composite with 20 wt.% reinforcement. The mechanical properties of the obtained specimens are presented in Table 5. As observed, the increase in the injection speed led to

Table 4. Tensile properties of flax/PP composites. (Impact type = notched Charpy Impact test).

Compounding method	Polymer	Flax fiber (wt.%)	Testing Temp. (°C)	Tensile strength (MPa)	Young's modulus (GPa)	Impact strength (kJ/m^2)
n/a	PP	0	RT	32.00 ± 1.00	2.0 ± 0.16	1.81 ± 0.35
	MA-PP (3 wt.%)	0	RT	34.50 ± 1.00	2.0 ± 0.15	2.03 ± 0.14
Kneader	MA-PP (3 wt.%)	28%	RT	42.00 ± 0.35	6.2 ± 0.40	–
Henschel mixer	MA-PP (3 wt.%)	28%	−25	39.92 ± 1.40	4.5 ± 0.16	–
			RT + 80	32.00 ± 0.80	3.8 ± 0.45	
				25.35 ± 1.80	2.5 ± 0.37	
Twin screw extruder	PP	10%	−25	49.00 ± 0.21	4.2 ± 0.13	1.83 ± 0.28
			RT + 80	29.30 ± 0.36	2.1 ± 0.19	
				13.20 ± 0.07	0.58 ± 0.10	
	PP	20%	−25	45.70 ± 0.93	5.3 ± 0.13	1.88 ± 0.13
			RT + 80	28.80 ± 0.94	2.6 ± 0.16	
				13.30 ± 0.06	0.7 ± 0.15	
	MA-PP (3 wt.%)	28%	RT + 80	35.20 ± 0.27	4.2 ± 0.26	3.40 ± 0.14
				19.90 ± 0.79	1.6 ± 0.13	
LFT	PP	28%	RT	35.00 ± 0.80	5.0 ± 1.19	7.23 ± 0.87

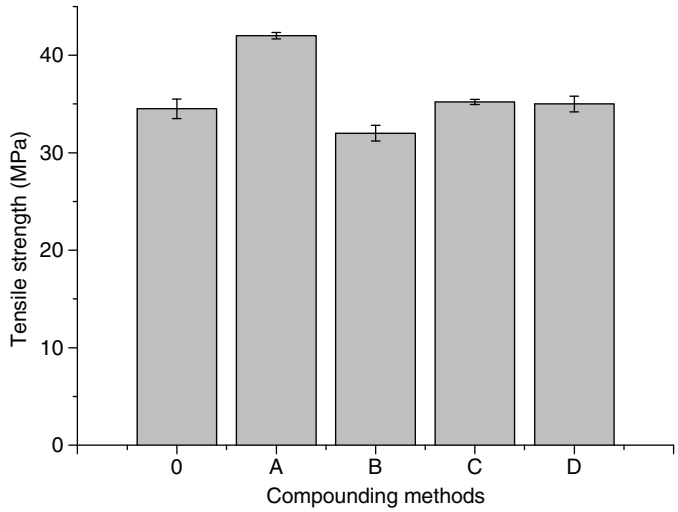

Figure 6. *Tensile strength of injection molded flax/PP composites compounded through various methods (28 wt.% flax fibers): (A) kneader, (B) Henschel mixer, (C) twin screw extruder and (D) LFT, (0) unreinforced MA-PP for reference.*

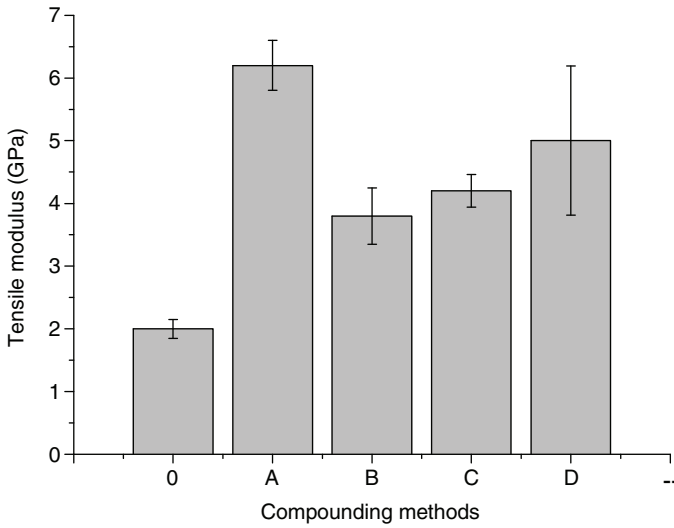

Figure 7. *Stiffness of injection molded flax/PP composites compounded through various methods (28 wt.% flax fibers): (A) kneader, (B) Henschel mixer, (C) twin screw extruder, (D) LFT, (0) unreinforced MA-PP for reference.*

minor improvement in the tensile properties. Such improvements in the tensile properties could be due to the improved fiber orientation along the direction of polymer flow and hence along the tensile bars. The aforementioned effect is more obvious for the stiffness of the composites rather than for the strength where the influence is minimal.

Thorough analysis on the influence of fiber length and concentration on the stiffness and strength of GF/PP [19,20] and flax/PP composites [18] has shown that the required fiber length to reach maximum stiffness is much lower than that required to

Table 5. Effect of injection rate during injection molding on the tensile properties of flax/PP composite (20 wt.% flax fibers).

Injection speed (cm^3/s)	Tensile strength (MPa)	Young's modulus (GPa)
11	28.74 ± 0.23	2.34 ± 0.80
18	28.97 ± 0.14	2.47 ± 0.23
26	28.77 ± 0.09	2.63 ± 0.16
35	29.50 ± 0.11	2.80 ± 0.28
44	29.71 ± 0.60	2.80 ± 0.32

maximum strength. More specifically, in the case of GF/PP, fiber lengths higher than 0.8 mm are already enough to create a short fiber composite with stiffness equivalent to 90% of that of a continuous fiber composite, while 90% strength levels are only obtained at fiber lengths as high as 13 mm. This means that the fibers need to be approximately 15 times longer to effectively increase the strength compared with the length needed for stiffness improvement of GF/PP composites. A similar analysis on flax/PP composites data [18] showed that the required fiber length for optimum stiffness is approximately 1 mm, while the respective length for optimum strength is approximately 8 mm (8 times higher fiber length for strength improvement). Based on this analysis, a better understanding of the data presented in Figures 6 and 7 is possible. The significant improvement in stiffness of composites obtained through all compounding methods is due to the fact that stiffness is less sensitive to fiber length compared to strength. As shown in Figure 3, in the case of kneading, highest fiber lengths are being obtained compared to Henschel mixer and twin screw extrusion compounding, therefore the highest stiffness improvement. It is interesting to note that next to kneading, the LFT composites had the second best improvement, even if no compatibilizer is used. Since the obtained fiber length is much higher than that required for stiffness improvement, no major effect was expected due to fiber–matrix adhesion. This is in line with previous observations on the effect of compatibilizer on the stiffness of flax/PP composites [18].

More quantitative information is required to analyze the effect of fiber length on the strength of the various composites. This information can be gained if we compare the critical fiber length for strength with the fiber lengths obtained through compounding and injection molding. For this, a similar approach is being followed with that presented in [2,5,16,18,21,22,31]. The critical fiber length (L_c) can be calculated using the Kelly-Tyson theory [41]:

$$L_c = \frac{\sigma_f d}{2\tau} \tag{1}$$

where σ_f is the fiber strength, d is the fiber diameter, and τ is the interfacial bond strength. For the critical fiber length calculations, the following data are being used based on literature: the strength of the technical fiber is 800 MPa [6,21], while that of an elementary fiber is 1500 MPa [6,21], for fibers with 80 and 12.5 μm diameter, respectively. Furthermore, interfacial bond strengths (τ) of 8 MPa for flax/PP [2,38,42,43] and 16 MPa [2] for flax/MA-PP, are being assumed. Using these values, the critical fiber length for technical and elementary flax fibers can be calculated and is presented in Table 6. Based on these data a critical fiber length of approximately 2 mm can be found in the case

Table 6. Critical fiber lengths for flax/PP and flax/MA-PP systems with elementary and technical fibers.

Critical fiber length	Flax/PP	Flax/MA-PP
Elementary fiber	1.2 mm	0.6 mm
Technical fiber	4.0 mm	2.0 mm

of flax/MA-PP with technical flax fibers. The analysis of the data in Figure 3 showed that only 5–12% of the fibers possessed lengths above the critical length when compounding methods A, B and C were followed. However, to attain short fiber composite strengths which are equivalent to 90% of tensile strength of a continuous fiber composite, fiber lengths $>5L_c$ are required [20]. This means that for effective strength improvement fiber lengths of approximately 10 mm are required for flax/MA-PP and of approximately 15 mm for flax/PP. These results are in agreement with [18] where optimum strength for flax/MA-PP is obtained for approximately 8 mm fiber lengths. From the fiber length distribution analysis in Figure 3, it is obvious that none of the compounding techniques allowed for such high fiber lengths. However, in the case of kneading, significant improvement in strength can be found. This behavior could be attributed to the fact that, as mentioned before, next to fiber breakage, compounding may also separate the technical cellulose fibers into finer elementary fibers [15], which can lead to higher fiber aspect ratios. If elementary fibers are assumed in combination with MA-PP matrix, then $5L_c$ equals to 3 mm. A considerable fraction of fibers is above 3 mm in the kneading process, which is not the case for the Henschel mixing or twin screw extrusion. Based on this assumption, it can be concluded that fiber separation was probably more pronounced during kneading than any other compounding method.

Effect of Compatibilization on the Mechanical Properties of Flax/PP

The influence of MA-PP concentration on composites, which were manufactured through injection molding (compounding through kneading), was investigated. The materials investigated consisted of composites with 28 wt.% of short flax fibers. As reference, a pure PP matrix was used. The tensile modulus of the injection-molded flax/MA-PP composites is given in Figure 8. From the figure, it can be seen that the modulus results show an approximately constant value of 6.2 and 1.7 GPa for the 28 wt.% series and the reference PP material, respectively. As expected, no influence of fiber–matrix adhesion on composite stiffness was observed.

The tensile strength values of the injection-molded flax/PP/MA-PP composites are given in Figure 9. The matrix tensile strength shows a constant value of around 30 MPa. On increasing the amount of MA-PP in the flax/PP/MA-PP composite, a maximum tensile strength was observed for 20 wt.% MA-PP. This behavior is in line with previous observation on the effect of MA-PP on the mechanical properties of GF/PP [44], where first an increase in strength with compatibilizer is found and a subsequent decrease at higher fractions of MA-PP. An optimum in interfacial shear strength was also observed for flax/MA-PP systems. However, here an optimum was found at 10 wt.% MA-PP content in micro-composites, measured through micro-debond tests [42]. The observed optimum in micro-debond tests is not directly reflected in the macro-composites properties, which can be because of effects related to fiber volume fractions (single fiber

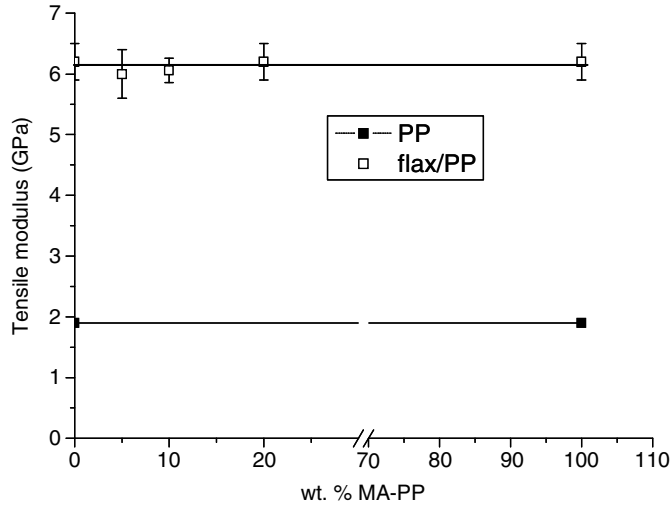

Figure 8. *Tensile modulus of injection molded flax/PP/MA-PP composites with varying amount of MA-PP content in the matrix (28 wt.% flax fibers).*

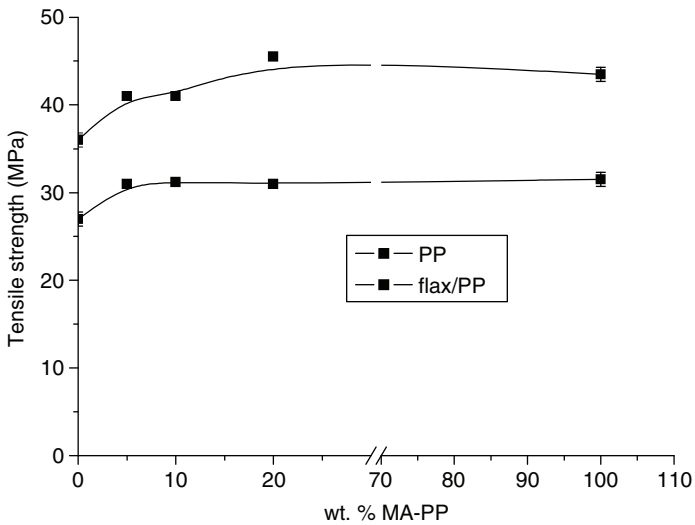

Figure 9. *Tensile strength of injection molded flax/PP/MA-PP composites with varying amount of MA-PP content in the matrix (28 wt.% flax fibers).*

versus real composite). In any case, such high weight fractions of MA-PP are not economical when a balance in price and property enhancement is desired. Such balance can however be obtained for 3–5 wt.% addition of MA-PP in PP.

Comparison of Injection Molding with Compression Molding Flax/PP

Figures 10 and 11 show the stiffness and strength of compression molded (NMT) [2] and injection-molded (compounding done through kneading) flax/3 wt.% MA-PP

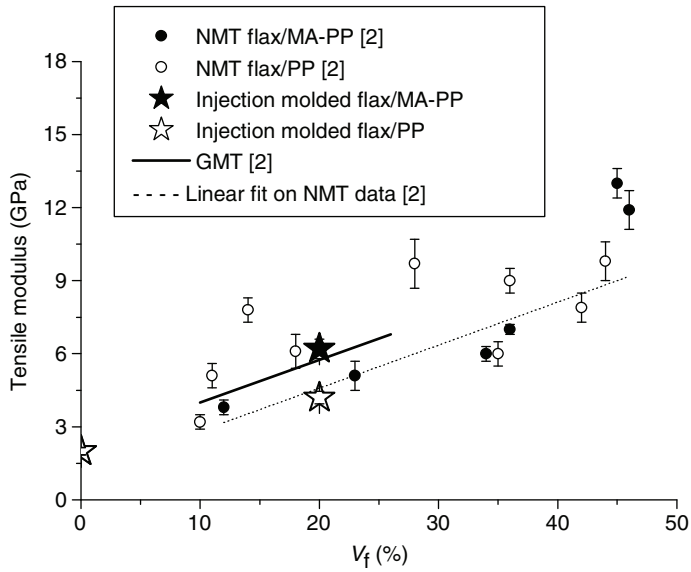

Figure 10. *Tensile modulus of flax/PP composites manufactured through compression molding of NMT [2] and injection molding as a function of fiber volume fraction. Also shown is data of GMT [2].*

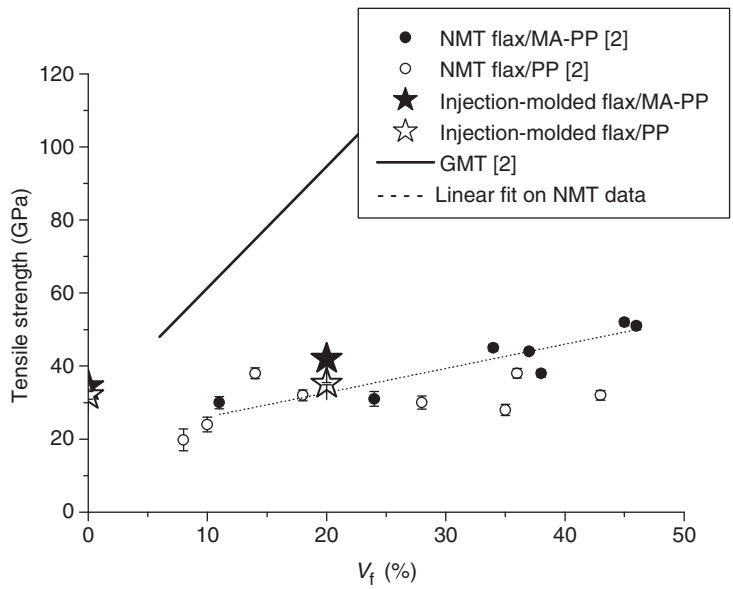

Figure 11. *Tensile strength of flax/PP composites manufactured through compression molding of NMT [2] and injection molding as a function of fiber volume fraction. Also shown is data of GMT [2].*

composites, respectively. In spite of a reduction in fiber length during injection molding, it can be seen that injection-molded samples showed tensile properties similar to that of NMT (fiber length ≈25 mm) compression molded composites. This could be because of reasons like: (a) improved fiber efficiency because of dimensional changes, i.e., reduction

in fiber diameter through fiber opening as discussed earlier and (b) changes in fiber orientation along the direction of polymer flow, in the case of injection-molded composites. Dimensional changes can play an important role since the flax fibers used for composite reinforcement are often actually fiber bundles (so-called technical fibers) of fiber cells (so-called elementary fibers) [2,38]. In addition, it has been shown that the fiber tensile strength is strongly dependent on the fiber length [21]; therefore, a reduction in fiber length could have led to separation of the fiber bundles to fiber cells or splitting hence leading to improved fiber efficiency through enhanced fiber tensile properties (of fiber cells) or improved fiber aspect ratio.

CONCLUSIONS

Compounding of flax with PP was carried out by four different methods: (i) kneader, (ii) Henschel kinetic mixer, (iii) fiber pelletizing followed by twin screw extrusion and (iv) production of LFTs through pultrusion. The problem of fiber feeding, in the case of fluffy flax fibers can be solved through the application of the Amandus Kahl pelletizing method. The fiber pelletizing method is cheaper than the long fiber granulation method and also more uniform granules are obtained when compared to kneading and Henschel kinetic mixer. From the analysis of the fiber length distribution in these compressed fiber pellets as well as compounded material, it was found that considerable fiber breakage occurs in the compounding stage in the case of kinetic mixing and extrusion.

Fiber addition through fiber pills followed by extrusion compounding led to a reduction in tensile strength when compared to pure PP. However, the addition of a compatibilizer (MA-PP) led to some improvements in tensile properties. Owing to higher fiber lengths in the LFT method, increased composite toughness was observed. Kneader compounded composites showed maximum tensile strength as well as stiffness when compared with other compounding methods. This could be because of higher fiber length retention, as observed in the fiber length distribution, as well as better fiber–matrix adhesion in the system studied.

Based on a comparative study between NMT compression molded and injection-molded flax/PP composites, it can be concluded that the reduction in fiber length associated with injection molding did not affect the tensile properties significantly. In fact, the reduction in fiber length here was counterbalanced by improvements in fiber orientation along the direction of polymer flow and fiber efficiency through improved fiber opening.

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